OPEN ACCESS
IOP Publishing
Journal of Optics
J. Opt. 18 (2016) 015404 (3pp)
doi:10.1088/2040-8978/18/1/015404

Energy conservation and the constitutive relations in chiral and non-reciprocal media

Stephen M Barnett and Robert P Cameron

School of Physics and Astronomy, University of Glasgow, Glasgow G12 8QQ, UK

E-mail: stephen.barnett@glasgow.ac.uk

Received 17 August 2015, revised 15 September 2015
Accepted for publication 17 September 2015
Published 11 December 2015

Abstract

We consider the possibility that the chirality parameters and the non-reciprocity parameters appearing in the constitutive relations for the displacement and magnetic induction fields in a bi-isotropic medium might not be equal and thereby shed light on the physical significance of the fact that they are the same. We find, in particular, that they must be equal in order to retain the local conservation of energy.

Keywords: chirality, optical activity, constitutive relations

Chiral molecules and chiral media reveal their nature through their interactions with polarized light. At the molecular level these arise from processes that include both magnetic-dipole and electric-quadrupole transitions together with the typically far stronger electric-dipole transitions [1, 2]. If the molecules are not aligned, however, then orientational averaging leads to a simpler description in which electric-quadrupole effects do not contribute. For such isotropic configurations it often suffices to replace the familiar consitutive relations for dielectric and magneto-dielectric media with the Drude–Born–Fedorov relations [3–5]

$$\begin{aligned} \mathbf{D} &= \varepsilon(\mathbf{E} + \beta\, \boldsymbol{\nabla} \times \mathbf{E}) \\ \mathbf{B} &= \mu(\mathbf{H} + \beta\, \boldsymbol{\nabla} \times \mathbf{H}). \end{aligned} \tag{1}$$

Here ε and μ are the familiar permittivity and permeability for the medium and β is the chirality parameter. We note that the chirality parameter is the same in both constitutive relations, those for **D** and for **B**. It is this similarity that concerns us in this short paper.

The symmetrical placing of the chirality parameter in the constitutive relations was not present in the earliest theories; in the work of Drude, for example, we find what is essentially the form [6]

$$\begin{aligned} \mathbf{D} &= \varepsilon(\mathbf{E} + \beta\, \boldsymbol{\nabla} \times \mathbf{E}) \\ \mathbf{B} &= \mu\mathbf{H}. \end{aligned} \tag{2}$$

There are fundamental reasons and, indeed, experimental evidence for favouring the forms (1) over (2). To these we add, in this paper, the observation that the fundamental requirements of the local and global conservation of energy require the form (1) with the chirality parameters equal. Our approach is to postulate the generalized constitutive relations

$$\begin{aligned} \mathbf{D} &= \varepsilon\left(\mathbf{E} + \beta_d\, \boldsymbol{\nabla} \times \mathbf{E}\right) \\ \mathbf{B} &= \mu\left(\mathbf{H} + \beta_b\, \boldsymbol{\nabla} \times \mathbf{H}\right) \end{aligned} \tag{3}$$

and show that energy is not conserved unless $\beta_d = \beta_b$.

The parameters appearing in the constitutive relations are, in general, frequency-dependent so that the complete expression of the constitutive relations has the form:

$$\begin{aligned} \mathbf{D}(\omega) &= \varepsilon(\omega)[\mathbf{E}(\omega) + \mathrm{i}\omega\beta(\omega)\mathbf{B}(\omega)] \\ \mathbf{B}(\omega) &= \mu(\omega)[\mathbf{H}(\omega) - \mathrm{i}\omega\beta(\omega)\mathbf{D}(\omega)]. \end{aligned} \tag{4}$$

These simple relations have been employed in the study of chiral optical media, their associated fields and forces on chiral objects [7–9] and they appear naturally in the study of bi-isotropic media [10, 11]. The constitutive relations are also sometimes written in the time domain in the form [12]

$$\begin{aligned} \mathbf{D} &= \varepsilon\left(\mathbf{E} - \beta\dot{\mathbf{B}}\right) \\ \mathbf{B} &= \mu\left(\mathbf{H} + \beta\dot{\mathbf{D}}\right), \end{aligned} \tag{5}$$

where it is understood that these apply only if the fields are restricted to a sufficiently narrow frequency bandwidth so that the medium properties, $\varepsilon\mu$ and β, may be approximated by constants. We note that similar expressions occur in some of the early theoretical work on this topic [13–15]. Brief but very readable accounts of the rival forms of the constitutive relations and the development of these may be found in the texts by Lakhtakia and his colleagues [3, 4].

The constitutive relations (1) possess a number of important symmetries. Principal among these are the properties under inversion in space or time and the Heaviside–Larmor or duality symmetry [16, 17].

A vector field, such as the electric field changes sign under spatial inversion, that is it is odd under the parity transformation, but a pseudo-vector field, like the magnetic field is unchanged and so is parity even. The curl of a vector field is a psuedo-vector field and that of a pseudo-vector field is a vector field. It necessarily follows that the chirality parameter, β, is a pseudo-scalar which changes sign under spatial inversion. This is entirely natural, of course, as this change under spatial reflection is the very nature of chirality. It follows directly from Maxwell's equations that the electric and magnetic fields have opposite properties under time reversal and if we agree that charges do not change sign under temporal inversion, then the electric field is unchanged under time reversal but the magnetic field changes sign. It is for this reason that the time-derivatives of $\mathbf{B}$ and $\mathbf{D}$ appear in the constitutive relations when written in the form (5) [16].

The form of the macroscopic Maxwell equations in the absence of free charges and currents exhibits an electric-magnetic symmetry or duality first expressed by Heaviside and Larmor [18, 19]. This symmetry, which is associated with the conservation of helicity [20], is most naturally expressed as an invariance under the duality transformation [12]:

$$\begin{aligned}
\mathbf{E} &\rightarrow \mathbf{E}\cos\xi + \sqrt{\frac{\mu}{\varepsilon}}\mathbf{H}\sin\xi \\
\mathbf{H} &\rightarrow -\sqrt{\frac{\varepsilon}{\mu}}\mathbf{E}\sin\xi + \mathbf{H}\cos\xi \\
\mathbf{D} &\rightarrow \mathbf{D}\cos\xi + \sqrt{\frac{\varepsilon}{\mu}}\mathbf{B}\sin\xi \\
\mathbf{B} &\rightarrow -\sqrt{\frac{\mu}{\varepsilon}}\mathbf{D}\sin\xi + \mathbf{B}\cos\xi
\end{aligned} \tag{6}$$

for any choice of the pseudoscalar ξ. We note that the constitutive relations (1) are invariant under this transformation and so respect the Heaviside–Larmor symmetry [17]. This is not true for the Drude form (2), nor for our postulated form (3) and this feature alone is a strong evidence in favour of (1). Further evidence, as we now show, comes from consideration of energy conservation for light propagating through a chiral medium.

It is known that the constitutive relations (1) are consistent with the local conservation of energy as expressed in the form a continuity equation [3, 13]. Our task is to show that energy is not conserved if we adopt an imbalanced form as in (3) or, as a special case, the Drude form (2). It is simplest to work with the form of the constitutive relations (5) in which the time derivatives appear, remembering that we have the implicit condition that the frequency bandwidth is sufficiently narrow that the medium parameters may be considered to be constant. We note also that chiroptical effects are typically small and it suffices to work to first order in the chirality parameter [13]. If we adopt the generalized constitutive relation (3) then we find that the familiar Poynting theorem for the local conservation of energy becomes

$$\begin{aligned}
&\frac{\partial}{\partial t}\frac{1}{2}(\mathbf{D}\cdot\mathbf{E} + \mathbf{B}\cdot\mathbf{H}) + \boldsymbol{\nabla}\cdot(\mathbf{E}\times\mathbf{H}) \\
&= \frac{1}{2}\epsilon\mu\left(\beta_b - \beta_d\right)\dot{\mathbf{E}}\cdot\dot{\mathbf{H}} \\
&\quad + \frac{1}{2}\epsilon\mu\left(\beta_d\mathbf{E}\cdot\ddot{\mathbf{H}} - \beta_b\ddot{\mathbf{E}}\cdot\mathbf{H}\right).
\end{aligned} \tag{7}$$

This has the familiar form of a local conservation law with a time derivative of a density, the divergence of a flux and what appears to be a source term, the latter appearing on the right-hand side of the equation. We need to be careful, however, to extract from this residual term any component that may be incorporated into the characteristic form of the continuity equation on the left-hand side of the equation. To this end we note that

$$\begin{aligned}
&\beta_d\mathbf{E}\cdot\ddot{\mathbf{H}} - \beta_b\ddot{\mathbf{E}}\cdot\mathbf{H} \\
&\quad = \frac{\partial}{\partial t}\frac{1}{2}\left(\beta_d + \beta_b\right)\left(\mathbf{E}\cdot\dot{\mathbf{H}} - \dot{\mathbf{E}}\cdot\mathbf{H}\right) \\
&\qquad + \frac{1}{2}\left(\beta_d - \beta_b\right)\left(\mathbf{E}\cdot\ddot{\mathbf{H}} + \ddot{\mathbf{E}}\cdot\mathbf{H}\right).
\end{aligned} \tag{8}$$

We can associate the first of these terms with a chiroptical contribution to the energy density but no similar interpretation is possible for the second term. Hence we can rewrite (7) in the form

$$\frac{\partial}{\partial t}w + \boldsymbol{\nabla}\cdot\mathbf{S} = \frac{1}{4}\epsilon\mu\left(\beta_d - \beta_b\right)\left(\mathbf{E}\cdot\ddot{\mathbf{H}} + \ddot{\mathbf{E}}\cdot\mathbf{H} - 2\dot{\mathbf{E}}\cdot\dot{\mathbf{H}}\right), \tag{9}$$

where w is the full energy density and $\mathbf{S}$ is the familiar Poynting vector:

$$\begin{aligned}
w &= \frac{1}{2}\left[\mathbf{D}\cdot\mathbf{E} + \mathbf{B}\cdot\mathbf{H} + \epsilon\mu\left(\beta_d + \beta_b\right)\left(\mathbf{E}\cdot\dot{\mathbf{H}} - \dot{\mathbf{E}}\cdot\mathbf{H}\right)\right] \\
\mathbf{S} &= \mathbf{E}\times\mathbf{H}.
\end{aligned} \tag{10}$$

It is clear that there is a source or sink term for the electromagnetic energy in the continuity equation (9) and this *cannot* be incorporated, in general, into the left-hand side of the equation as the time derivative of an energy density. We may conclude that the energy of an electromagnetic field propagating through a non-absorbing chiral medium characterized by the generalized constitutive relation (3) leads to a *violation* of the local conservation of energy unless, of course, the two chiral parameters, β_d and β_b are precisely equal, which is the currently accepted form as given in (1).

Such observations are readily extended to a general bi-isotropic medium. To illustrate this, let us now postulate the constitutive relations

$$\begin{aligned}
\mathbf{D} &= \varepsilon(\mathbf{E} + \beta\,\boldsymbol{\nabla}\times\mathbf{E}) + \alpha_d\mathbf{H} \\
\mathbf{B} &= \mu(\mathbf{H} + \beta\,\boldsymbol{\nabla}\times\mathbf{H}) + \alpha_b\mathbf{E},
\end{aligned} \tag{11}$$

where we have allowed here for the possibility that the Tellegen or non-reciprocity parameters [21, 22] α_d and α_b are not equal, in which case electric-magnetic duality is not respected. We then find that

$$\frac{\partial}{\partial t}w + \boldsymbol{\nabla}\cdot\mathbf{S} = \frac{1}{2}(\alpha_d - \alpha_b)\big(\dot{\mathbf{E}}\cdot\mathbf{H} - \mathbf{E}\cdot\dot{\mathbf{H}}\big) \tag{12}$$

to first order in the chirality parameter and non-reciprocity parameters. Evidently, the local conservation of energy [22] is only ensured provided $\alpha_d = \alpha_b$, as is the case, of course, in reality. It is interesting to note that the quantity $(\dot{\mathbf{E}}\cdot\mathbf{H} - \mathbf{E}\cdot\dot{\mathbf{H}})/2$ which appears in (10) and (12) is itself a density of a conserved property of freely propagating light; the 00-zilch [23, 24].

That the chirality parameters and the non-reciprocity parameters appearing in the constitutive relations for **D** and **B** in a bi-isotropic medium are equal is now well established and this fact may be traced back to fundamental quantum mechanical interaction of atoms and molecules with light [2]. It is also clear that this equality is required by the more subtle nature of the duality symmetry of Heaviside and Larmor [17]. To these convincing arguments we can now add the observation that this equality is also required by the local conservation of energy in an electromagnetic wave propagating through a bi-isotropic medium. It is, perhaps, this requirement that provides the simplest and most fundamental reason for the equalities.

Acknowledgments

This work was supported by the UK Engineering and Physical Sciences Research Council under grant numbers EP/I012451/1 and EP/M004694/1. We thank an anonymous referee who brought to our attention the presence of a 00-zilch density in our results and whose questions inspired us to extend our analysis to general bi-isotropic media.

References

[1] Craig D P and Thirunamachandran T 1984 *Molecular Quantum Electrodynamics* (London: Academic)
[2] Barron L D 2004 *Molecular Light Scattering and Optical Activity* 2nd edn (Cambridge: Cambridge University Press)
[3] Lakhtakia A, Varadan V K and Varadan V V 1989 *Time-Harmonic Electromagnetic Fields in Chiral Media* (Berlin: Springer)
[4] Lakhtakia Beltrami A 1994 *Fields in Chiral Media* (Singapore: World Scientific)
[5] Bohren C F 2003 Isotropic chiral materials *Introduction to Complex Mediums for Optics and Electromagnetics* ed W S Weiglhofer and A Lakhtakia (Bellingham, WA: SPIE Digital Library)
[6] Drude P 1933 *The Theory of Optics* (London: Longmans Green) This is a translation into English by Riborg Mann C and Millikan R A 1900 of the original 1900 text Lehrbuch der Optik
[7] Marqués R, Medina F and Rafii-El-Idrissi R 2002 Role of bianisotropy in negative permeability and left-handed metamaterials *Phys. Rev.* B **65** 144440
[8] Canaguier-Durand A, Hutchison J A, Genet C and Ebbesen T W 2013 Mechanical separation of chiral dipoles by chiral light *New J. Phys.* **15** 123037
[9] Alizadeh M H and Reinhard B M 2015 Plasmonically enhanced chiral optical fields and forces in achiral split ring resonators *ACS Photonics* **2** 361–8
[10] Lakhtakia A and Weiglhofer W S 1995 On light propagation in helicoidal bianisotropic mediums *Proc. R. Soc. London* A **448** 419–37
[11] Weiglhofer W S 2003 Constitutive characterization of simple and complex mediums *Introduction to Complex Mediums for Optics and Electromagnetics* ed W S Weiglhofer and A Lakhtakia (Bellingham, WA: SPIE Digital Library)
[12] Jackson J D 1999 *Classical Electrodynamics* 3rd edn (New York: Wiley)
[13] Bursian V and Timorew A 1926 Zur theorie der optisch isotropen medien *Z. Phys.* **38** 475–84
[14] Rosenfeld L 1928 Quantenmechanische theorie der natürlichen optischen aktivität von Flüssigkeiten und gasen *Z. Phys.* **52** 161–74
[15] Condon E U 1937 Theories of optical rotary power *Rev. Mod. Phys.* **9** 432–57
[16] Satten R A 1958 Time-reversalsymmetry and electromagnetic polarization conditions *J. Chem. Phys.* **28** 742–3
[17] Silverman M P 1986 Reflection and refraction at the surface of a chiral medium: comparison of gyrotropic constitutive relations invariant or non-invariant under a duality transformation *J. Opt. Soc. Am.* A **3** 830–7
[18] Heaviside O 1892 On the forces, stresses and fluxes of energy in the electromagnetic field *Phil. Trans. R. Soc.* A **183** 423–80
[19] Larmor J 1897 Dynamical theory of the electric and luminiferous medium: III. *Phil. Trans. R. Soc.* A **190** 205–300
[20] Barnett S M, Cameron R P and Yao A M 2012 Duplex symmetry and its relation to the conservation of optical helicity *Phys. Rev.* A **86** 013845
[21] Tellegen B D F 1948 The gyrator, a new electric network element *Phillips Research Report* 3 pp 81–101
[22] Altan B S 2008 A uniqueness theorem for initial-boundary value problems in Tellegen medium *Prog. Electromagn. Res.* C **1** 173–85
[23] Lipkin D M 1964 Existence of a new conservation law in electromagnetic theory *J. Math. Phys.* **5** 696–700
[24] Ragusa S 1994 Electromagnetic first-order conservation laws in a chiral medium *J. Phys. A: Math. Gen.* **27** 2887–90